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TITLE: KINETIC STUDIES FOR XeF and KrF LASERS

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The goal of this program is to measure both the radiative lifetime and the collisional quenching rate coefficients for the XeF and KrF upper laser levels. Our approach is to excite a fraction of the ground state molecules in a cell using a short-pulse XeF or KrF laser. The ground state molecules will be prepared by flash dissociating a mixture of F_2 and either Xe or Kr plus a monatomic diluent and then allowing the molecular XeF to come to equilibrium with the free atom densities. This approach has the advantage over discharge and electron beam techniques that there will be no residual excited atomic states formed that can interfere with the subsequent fluorescence decay measurement.

A schematic diagram showing the experimental layout is presented in Figure 1. The flash photolysis cell and the associated gas manifold have been designed and assembled. The photolysis cell is constructed of quartz with four UV grade quartz windows in a cross configuration. The cell will be evacuated with an oil diffusion pump equipped with an anti-creep, liquid nitrogen cooled baffle. The cell is connected to the diffusion pump via a high-purity stainless manifold which will be passivated with F_2 before use. The entire vacuum system has been assembled and is currently being checked for leaks.

Gas mixtures for the experiment will be made up on a separate diffusion pumped manifold and then transferred to the photolysis cell manifold in stainless steel bottles equipped with demountable high vacuum fittings. This high purity manifold, which was previously used for preparing XeF and KrF laser gas mixtures, has been enlarged so that up to six gas mixtures can be prepared simultaneously. It has also been equipped with demountable high vacuum fittings utilizing Teflon gaskets so that the bottles containing the mixtures to be photolyzed can be removed and transferred to the high vacuum manifold connected to the photolysis cell.

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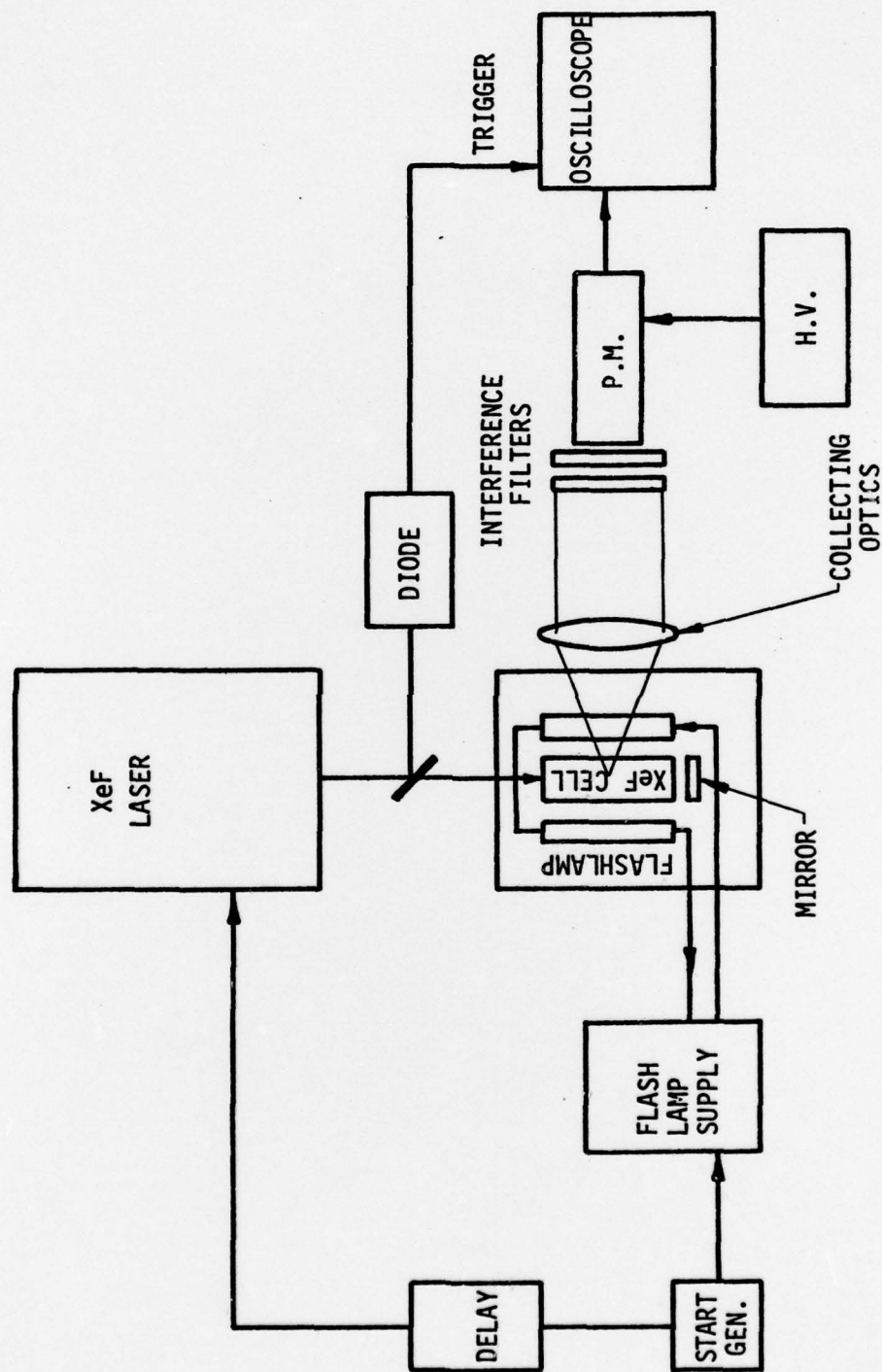


Figure 1. Schematic Diagram of the Laser Induced Fluorescence Experiment

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The inertness of the manifold was checked by making up Ar + F₂ mixtures and then verifying the nominal F₂ concentration using vacuum ultraviolet absorption.

The flashlamps chosen for these experiments (Xenon N-734C) have spectral distributions which typically peak in the region of the F₂ continuous absorption band at 2800 Å. For optically thin conditions, which will apply in this experiment, the dissociation fraction $[F]/2[F_2]$ is independent of the F₂ concentration and is given by

$$\frac{1}{2} \frac{[F]}{[F_2]} = \int \sigma(\nu) \phi(\nu) d\nu$$

where $\sigma(\nu)$ is the F₂ absorption cross section and $\phi(\nu)$ is the time-integrated photon flux from the flashlamp. According to the manufacturer, approximately 10 percent of the electrical energy input to the lamp is radiated as light in the 2500 to 3500 Å region. Assuming an average F₂ absorption cross section of 10⁻²⁰ cm² and an input energy of 100 J into a lamp with 7.5-cm arc length, then the dissociation fraction will be approximately 2 percent.

Maximum coupling between the photolysis cell and the flashlamps will be accomplished by locating them inside a double ellipsoidal reflector with the cell positioned on the common axis. The flashlamps are to be connected in series electrically and will be triggered by applying a voltage higher than the anode to cathode breakdown potential. The power supply consists of a 2 μF capacitor controlled by a triggered spark gap and is capable of providing 50 J to each lamp for a charge voltage of 10 kV. Care has been taken in the design of the flashlamp circuit to minimize the inductance in order to maximize the UV light output. The capacitor, spark gap, and cable assembly contribute approximately 125 nH to the circuit inductance, while the loop containing the two flashlamps in series contributes about 300 nH. This results in a circuit time constant $\tau = \sqrt{LC} \sim 1 \mu\text{sec}$, which should be about one third of the electrical pulse width. The mounting hardware for the flashtubes and the photolysis cell has been fabricated and is presently

being installed in the reflector and connected to the capacitor-spark-gap assembly.

The Blumlein-driven discharge laser has been equipped with a flash-board preionization circuit to allow operation at pressures above 1 atm. The UV pulse creates enough ionization in the gas to prevent arc formation at the higher pressures needed to obtain a short laser pulse width. Photodiode measurements indicate that the UV output from the flashboard reaches a maximum at approximately 200 to 300 nsec after initiation. Optimum performance of the XeF laser is obtained by firing the main discharge at the peak of the UV preionization pulse, although good performance is still obtained with longer delays. The output energy at 1 atm with a He + 1% Xe + 0.3% NF₃ gas mixture is typically 10 to 15 mJ. Laser pulse durations as short as 10 nsec have been obtained by carefully adjusting the experimental conditions. An oscilloscope trace showing the photodiode response for a XeF laser pulse is presented in Figure 2. This trace was obtained with a Blumlein charge voltage of 15 kV and a He + 1% Xe + 0.3% NF₃ gas mixture at 1.0 atm. The optical cavity consisted of a 2-m radius-of-curvature mirror with maximum reflectivity at 3500 Å opposite an uncoated fused-silica flat. The laser flux necessary to bleach the XeF absorption transition can be estimated from $\phi > \frac{h\nu}{\sigma\tau} \sim 75 \text{ kW/cm}^2$ for $\sigma\tau = 7.5 \times 10^{-25} \text{ sec-cm}^2$. (It should be noted that the product $\sigma\tau$ is independent of the XeF radiative lifetime and, therefore, so is the estimated laser flux.) The measured laser flux of several hundred kilowatts per square centimeter is considerably larger than this value and should be able to bleach the XeF absorption transition readily. However, it may be necessary to shorten the laser pulse width in order to observe the fastest decay rates.

Excessive electrical noise levels generated by the Blumlein laser discharge and rail switch are being picked up by our fast oscilloscope even though it is enclosed in a screen room. The Blumlein laser will be enclosed in a box constructed of two layers of brass screen in order to attenuate the noise to an acceptable level. This screen box is presently under construction and will be placed around the laser when it is completed.

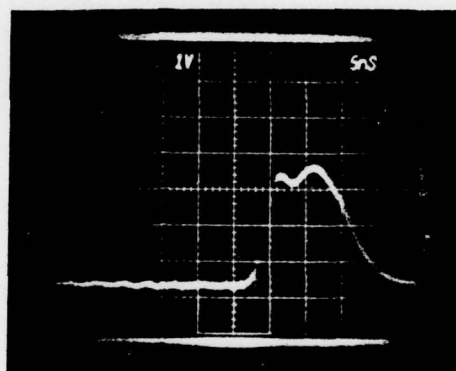


Figure 2. Photodiode Trace Showing Temporal History of XeF Laser Emission for a Blumlein Charge Voltage of 15 kV and a He + 1% Xe + 0.3% NF₃ Gas Mixture at 1 atm

It is anticipated that during the next quarter assembly and testing of the experimental system will be completed allowing preliminary lifetime data to be obtained for XeF.